On limitations of the Bruggeman formalism for inverse homogenization

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Abstract

The Bruggeman formalism provides an estimate ϵ_{hcm}^{Br} of the relative permittivity of a homogenized composite material (HCM), arising from two component materials with relative permittivities ϵ_a and ϵ_b. It can be inverted to provide an estimate of ϵ_a, from a knowledge of ϵ_{hcm}^{Br} and ϵ_b. Numerical studies show that the inverse Bruggeman estimate ϵ_a can be physically implausible when (i) $\text{Re}\left\{\epsilon_{hcm}^{Br}\right\}/\text{Re}\left\{\epsilon_b\right\} > 0$ and the degree of HCM dissipation is moderate or greater; or (ii) $\text{Re}\left\{\epsilon_{hcm}^{Br}\right\}/\ \text{Re}\left\{\epsilon_b\right\} < 0$ regardless of the degree of HCM dissipation. Furthermore, even when the inverse Bruggeman estimate is not obviously implausible, huge discrepancies can exist between this estimate and the corresponding estimate provided by the inverse Maxwell Garnett formalism.

Keywords: Bruggeman formalism; Maxwell Garnett formalism; inverse homogenization; metamaterials

1 Introduction

A composite material may be regarded as being effectively homogeneous, provided that wavelengths are much larger than the particle sizes of the component materials that make up the composite material. The constitutive parameters of such a homogenized composite material (HCM) can be estimated from a knowledge of the constitutive parameters of its component materials, along with a knowledge of the distributional statistics and shapes of its component particles [1, 2]. The Bruggeman homogenization formalism has been widely applied for this purpose for the past 70 years [1, 3]; and new areas of application for the Bruggeman formalism continue to emerge, for examples, in recent developments pertaining to complex HCMs [4, 5] and negatively–refracting metamaterials [2, 6]. However, a certain limitation of the Bruggeman homogenization formalism came to light in 2004 [7]. In the context of isotropic dielectric HCMs, derived from two component materials with relative permittivities ϵ_a and ϵ_b, it transpires that the Bruggeman estimate of the HCM relative permittivity may be physically implausible if $\epsilon_a/\epsilon_b < 0$ in the case of nondissipative HCMs or if $\text{Re}\left\{\epsilon_a\right\}/\text{Re}\left\{\epsilon_b\right\} < 0$ in the case of weakly dissipative HCMs. An example of such a problematic homogenization scenario — of considerable interest to the metamaterial community — arises in the homogenization of silver particles with insulating particles at visible and near infrared wavelengths [8]. This limitation — which is relevant to active [9] as well as nondissipative and dissipative HCMs — also extends to the Maxwell Garnett homogenization formalism which shares a common provenance with the Bruggeman formalism [10], as well as the Hashin–Shtrikman, Wiener and Bergman–Milton bounds on the HCM's relative permittivity [7, 11]. Similar anomalous results, for HCMs arising from two component materials with $\text{Re}\left\{\epsilon_a\right\}/\text{Re}\left\{\epsilon_b\right\} < 0$, have been described as 'electrostatic resonances' [12, 13, 14], but this term is avoided in [7, 8, 9, 11] (and herein) since the estimates of the HCM's relative permittivity described in [7, 8, 9, 11] are not physically plausible.

Restricting our attention to the simplest possible case of an isotropic dielectric HCM arising from two isotropic dielectric component materials, in this communication we investigate the applicability of the Brugge-

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man formalism to the inverse homogenization scenario wherein the relative permittivity of one of the component materials is estimated from a knowledge the relative permittivities of the other component material and the HCM. Formal expressions have been established for the inverse Bruggeman formalism (and the inverse Maxwell Garnett formalism) in the general setting of bianisotropic HCMs [15], but in certain cases these formal expressions may be ill–defined [16] and the ranges of applicability of these inverse formalisms have not been established. Our study is partly motivated by very recent implementations of the inverse Bruggeman formalism in estimating nanoscale constitutive and morphological parameters of certain sculptured thin films [17], which is a key step in modelling the electromagnetic response of infiltrated sculptured thin films [18, 19].

2 Analysis and numerical studies

We consider the homogenization of two isotropic dielectric component materials with relative permittivities ϵ_a and ϵ_b. The component materials a and b are assumed to be distributed randomly as spherical particles with volume fractions f_a and $f_b = 1 - f_a$, respectively. The Bruggeman estimate of the relative permittivity of the corresponding HCM, namely ϵ_{hcm}^{Br}, is provided via [3]

$$f_a \frac{\epsilon_a - \epsilon_{hcm}^{Br}}{\epsilon_a + 2\epsilon_{hcm}^{Br}} + f_b \frac{\epsilon_b - \epsilon_{hcm}^{Br}}{\epsilon_b + 2\epsilon_{hcm}^{Br}} = 0, \tag{1}$$

which is nonlinear in ϵ_{hcm}^{Br}. A straightforward manipulation of (1) delivers the explicit formula

$$\epsilon_a = \frac{(f_a - 2f_b)\,\epsilon_b + 2\epsilon_{hcm}^{Br}}{f_b\left(\epsilon_b - \epsilon_{hcm}^{Br}\right) + f_a\left(\epsilon_b + 2\epsilon_{hcm}^{Br}\right)}\,\epsilon_{hcm}^{Br} \tag{2}$$

for ϵ_a in terms of ϵ_b, ϵ_{hcm}^{Br}, f_a and f_b. Since the component materials a and b are treated in an identical manner within the Bruggeman formalism, the corresponding formula for ϵ_b has the same form as (2). Notice that as the inverse Bruggeman equation (2) does not involve a square root, there is no scope for $\mathrm{Im}\,\{\epsilon_a\}$ being nonzero if $\epsilon_b, \epsilon_{hcm}^{Br} \in \mathbb{R}$. This contrasts with the forward Bruggeman formalism where a square root term enables $\mathrm{Im}\left\{\epsilon_{hcm}^{Br}\right\}$ to be nonzero even though $\epsilon_a, \epsilon_b \in \mathbb{R}$. This physically–implausible scenario can arise when $\epsilon_a/\epsilon_b < 0$ [7].

For comparison, we introduce the Maxwell Garnett estimate of the HCM relative permittivity [3]

$$\epsilon_{hcm}^{MG} = \epsilon_b + \frac{3f_a\epsilon_b\,(\epsilon_a - \epsilon_b)}{\epsilon_a + 2\epsilon_b - f_a\,(\epsilon_a - \epsilon_b)} \tag{3}$$

and its corresponding inverse

$$\epsilon_a = \frac{(2 + f_a)\,\epsilon_{hcm}^{MG} - 2f_b\epsilon_b}{(1 + 2f_a)\,\epsilon_b - f_b\epsilon_{hcm}^{MG}}\,\epsilon_b. \tag{4}$$

The limiting behaviour of the inverse Bruggeman estimate (2) as compared with that of the inverse Maxwell Garnett estimate (4) is especially revealing. In the limit $f_a \to 1$, both estimates yield the relative permittivity of the HCM, as they must.[2] In the limit $f_a \to 0$, the inverse Bruggeman formalism yields $\epsilon_a \to -2\epsilon_{hcm}^{Br}$ whereas the inverse Maxwell Garnett formalism yields $\epsilon_a \to -2\epsilon_b$. Therefore, the two inverse estimates differ markedly as f_a approaches zero, provided that ϵ_b and the relative permittivity of the HCM are sufficiently different.

We now explore the inverse Bruggeman estimate (2), in comparison with the inverse Maxwell Garnett estimate (4), by means of some illustrative numerical examples. For nondissipative scenarios, the forward Bruggeman formalism runs into difficulties when $\epsilon_a/\epsilon_b < 0$ but not when $\epsilon_a/\epsilon_b > 0$ [7]. Accordingly, let us

[2] The Maxwell Garnett estimate of the HCM relative permittivity is only strictly applicable in the dilute composite regime $f_a \lesssim 0.3$. Accordingly, estimates of ϵ_a delivered by the inverse Maxwell Garnett formalism are strictly valid only for $f_a \lesssim 0.3$. However, ϵ_{hcm}^{MG} coincides with one of the Hashin–Shtrikman bounds on the HCM relative permittivity which applies at all values of f_a [20].

begin by focussing on the regimes $\epsilon_{hcm}^{Br,MG}/\epsilon_b < 0$ and $\epsilon_{hcm}^{Br,MG}/\epsilon_b > 0$. In Fig. 1, plots of ϵ_a, as determined by the inverse Bruggeman formalism and the inverse Maxwell Garnett formalism, versus f_a are provided for the cases where $\epsilon_b = \pm 2$ and $\epsilon_{hcm}^{Br,MG} = 3$. When $\epsilon_{hcm}^{Br,MG}/\epsilon_b > 0$ the inverse Bruggeman and inverse Maxwell Garnett estimates are in fairly close agreement. However, the values of ϵ_a yielded by the two inverse formalisms differ markedly when $\epsilon_{hcm}^{Br,MG}/\epsilon_b < 0$, except in the limit as f_a approaches unity. Most notably, the inverse Bruggeman estimate becomes singular and undergoes a change in sign as the volume fraction increases through $f_a = 0.56$, whereas the inverse Maxwell Garnett value remains finite and does not change sign.

Next we turn to dissipative homogenization scenarios. In the case of the forward Bruggeman formalism, problems arise when $\mathrm{Re}\{\epsilon_a\}/\mathrm{Re}\{\epsilon_b\} < 0$ and the degree of dissipation is relatively small; if $\mathrm{Re}\{\epsilon_a\}/\mathrm{Re}\{\epsilon_b\} < 0$ and the degree of dissipation is relatively large or if $\mathrm{Re}\{\epsilon_a\}/\mathrm{Re}\{\epsilon_b\} > 0$ then the forward Bruggeman formalism was found to deliver physically plausible estimates of the HCM relative permittivity [7]. Accordingly, we consider the regimes where $\mathrm{Re}\left\{\epsilon_{hcm}^{Br,MG}\right\}/\mathrm{Re}\{\epsilon_b\} > 0$ with the degree of dissipation in the HCM being relatively small, moderate and large. Graphs of the real and imaginary parts of ϵ_a, as estimated by the inverse Bruggeman and inverse Maxwell Garnett formalisms, are plotted versus f_a in Fig. 2 for the cases $\epsilon_b = 2$ and $\epsilon_{hcm}^{Br,MG} = 3 + \delta i$ where $\delta \in \{0.1, 1, 10\}$. When the degree of HCM dissipation is relatively small ($\delta = 0.1$), the estimates of the real and imaginary parts of ϵ_a provided by the inverse Bruggeman and inverse Maxwell Garnett formalisms agree fairly closely. When the degree of HCM dissipation is moderate ($\delta = 1$), there is still fairly close agreement between the inverse Bruggeman and inverse Maxwell Garnett values of ϵ_a for most values of f_a. Crucially, however, for $f_a < 0.05$ the imaginary part of ϵ_a estimated by the inverse Bruggeman formalism is negative–valued (unlike $\mathrm{Im}\{\epsilon_a\}$ estimated by the inverse Maxwell Garnett formalism which is positive–valued). Here $\mathrm{Im}\{\epsilon_a\} < 0$ is not a physically plausible outcome as it implies that the homogenization of an active material a and a nondissipative material b results in a dissipative HCM. For both the real and imaginary parts of ϵ_a, the discrepancies between the values estimated by the two inverse formalisms become enormous when the degree of HCM dissipation is relatively large ($\delta = 10$). Furthermore, the inverse Bruggeman estimate is physically implausible for a much larger range of f_a values; i.e., $\mathrm{Im}\{\epsilon_a\}$ estimated by inverse Bruggeman formalism is negative–valued for $f_a < 0.3$ when $\delta = 10$.

Lastly, we explore the $\mathrm{Re}\left\{\epsilon_{hcm}^{Br,MG}\right\}/\mathrm{Re}\{\epsilon_b\} < 0$ regime. Plots of the real and imaginary values of ϵ_a in Fig. 3 correspond to the same parameter values as those used for Fig. 2 except that here $\epsilon_b = -2$. The estimates of the inverse Bruggeman formalism are now physically implausible — due to $\mathrm{Im}\{\epsilon_a\} < 0$ — for a wide range of f_a values, regardless of whether the degree of HCM dissipation is relatively small, moderate or large. In contrast, the estimate of $\mathrm{Im}\{\epsilon_a\}$ provided by the inverse Maxwell Garnett formalism is positive–valued for all scenarios considered. Additionally, the real parts of ϵ_a delivered by the two inverse formalisms differ enormously except when f_a approaches unity, for all degrees of HCM dissipation considered.

3 Closing remarks

In the case of dissipative HCMs, the inverse Bruggeman estimates of ϵ_a can be physically implausible when:

(i) $\mathrm{Re}\left\{\epsilon_{hcm}^{Br}\right\}/\mathrm{Re}\{\epsilon_b\} > 0$ and the degree of HCM dissipation is moderate or greater; or

(ii) $\mathrm{Re}\left\{\epsilon_{hcm}^{Br}\right\}/\mathrm{Re}\{\epsilon_b\} < 0$ regardless of the degree of HCM dissipation.

In the case of nondissipative HCMs, enormous discrepancies can exist between the estimates of ϵ_a provided by the inverse Bruggeman formalism and the inverse Maxwell Garnett formalism when $\epsilon_{hcm}^{Br,MG}/\epsilon_b < 0$. Therefore, the inverse Bruggeman formalism should be applied with great caution. Finally, we note that in the very recent implementations of the inverse Bruggeman formalism which motivated this study [17, 18, 19], the relative permittivity parameters were positive–valued and the materials were nondissipative. The estimates yielded by the inverse Bruggeman formalism in these cases seem physically plausible, but the acid test can only be provided by suitable experimental measurements.

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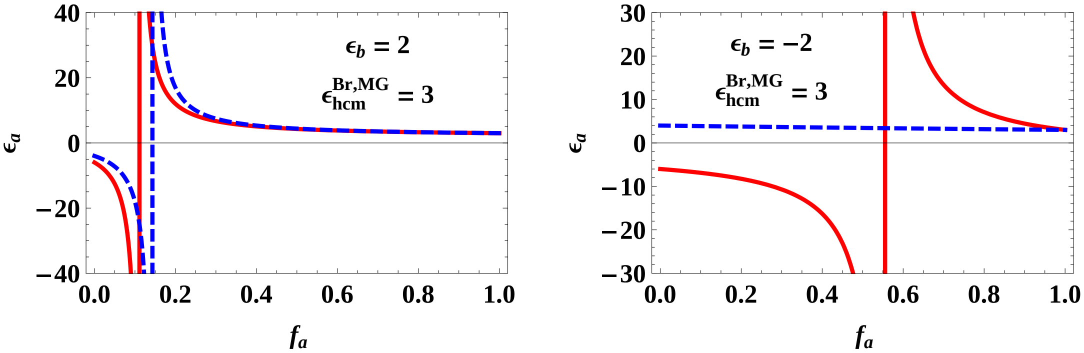

Figure 1: Plots of ϵ_a as determined by the inverse Bruggeman formalism (red, solid curves) and the inverse Maxwell Garnett formalism (blue, dashed curves) versus f_a for $\epsilon_b = \pm 2$ and $\epsilon_{hcm}^{Br,MG} = 3$. Estimates of ϵ_a delivered by the inverse Maxwell Garnett formalism are strictly valid only for $f_a \lesssim 0.3$.

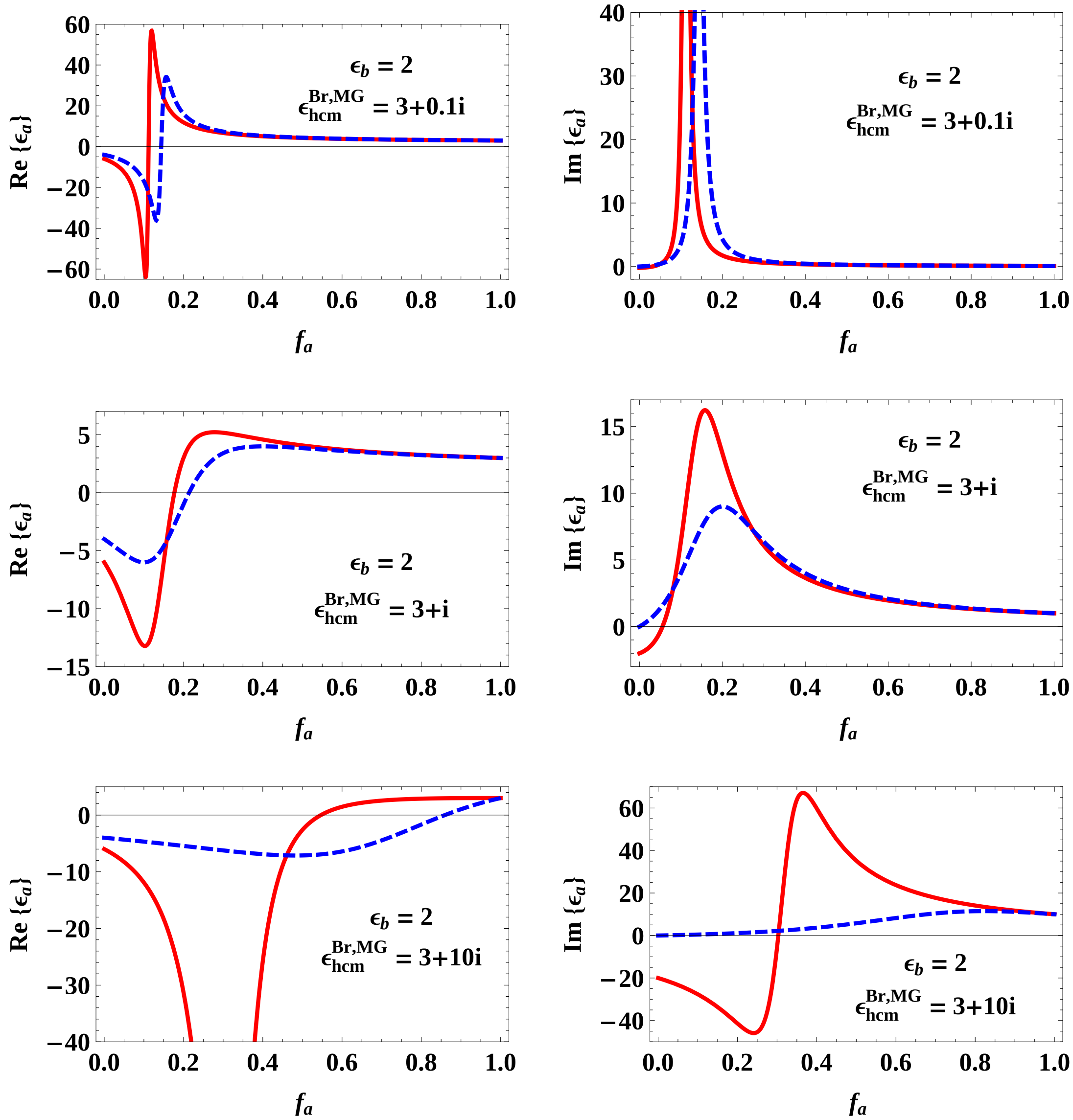

Figure 2: Plots of the real and imaginary parts of ϵ_a as determined by the inverse Bruggeman formalism (red, solid curves) and the inverse Maxwell Garnett formalism (blue, dashed curves) versus f_a for $\epsilon_b = 2$ and $\epsilon_{hcm}^{Br,MG} = 3 + \delta i$ where $\delta \in \{, 0.1, 1, 10\}$. Estimates of ϵ_a delivered by the inverse Maxwell Garnett formalism are strictly valid only for $f_a \lesssim 0.3$.

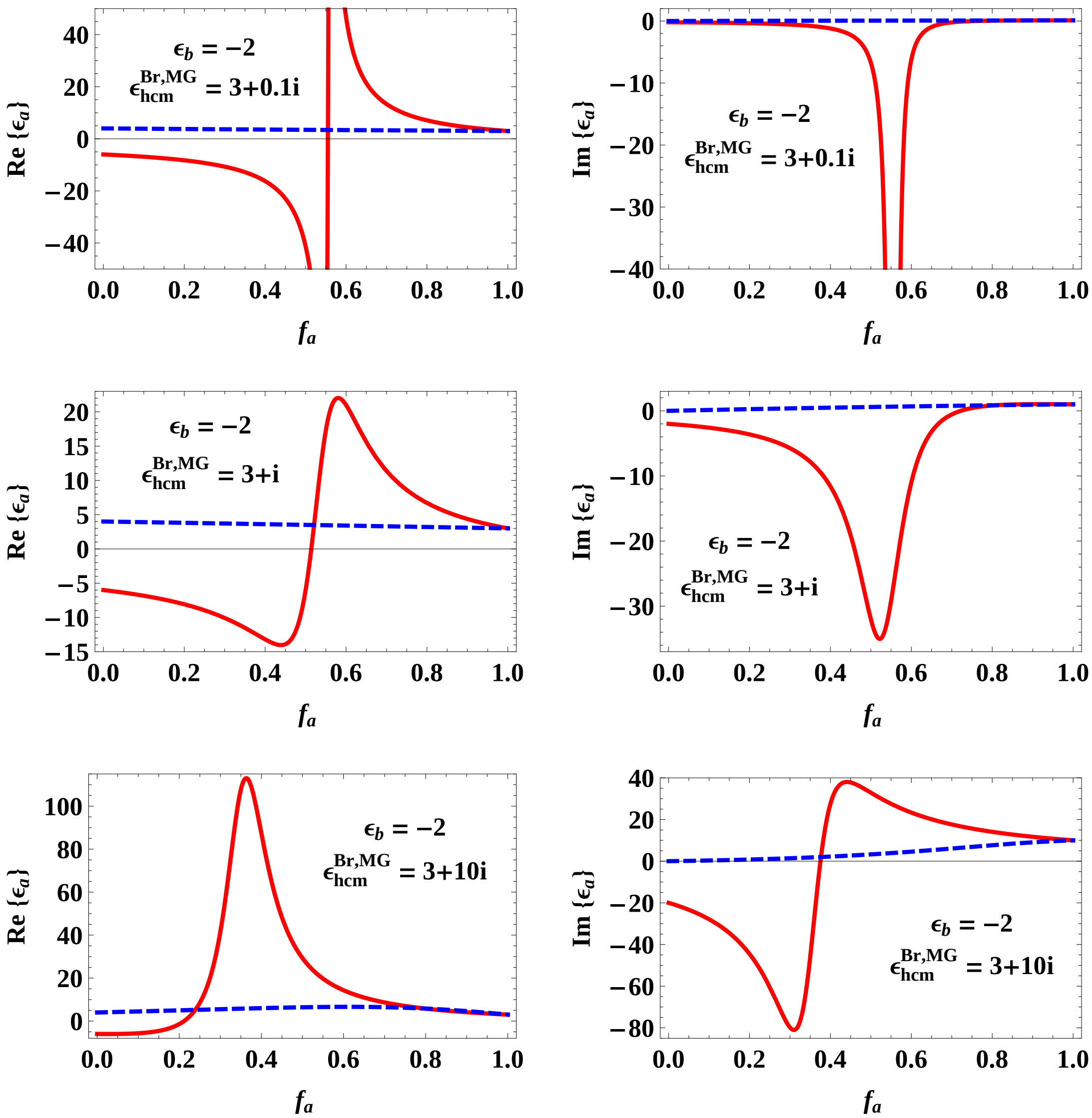

Figure 3: As Fig. 2 except that $\epsilon_b = -2$.